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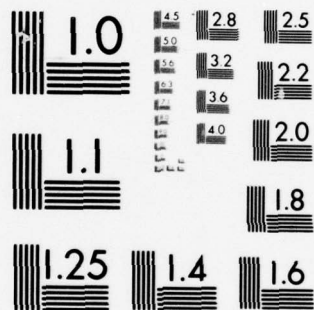

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FOURTH ORDER ELASTIC CONSTANTS FOR SODIUM CHLORIDE  
TO 270 KBAR BY WAVE DYNAMICS

Moayyed A. Hussain  
Julius Frankel  
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March 1978



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<p>HAVE BEEN DERIVED.</p> <p>We derive the dilatational and shear velocities and the equation of state for an isotropic material under high hydrostatic pressures by the methods of continuum mechanics using the theory of small deformation superposed on a finite strain. The strain energy density of the material is taken to fourth order in terms of the strain invariants.</p>		

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## TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
DERIVATION OF VELOCITIES FOR THE THEORY OF SMALL DEFORMATION SUPERPOSED ON FINITE UNIFORM DEFORMATION	2
The Deformed State (Under Finite Deformation)	5
The Perturbed State	6
THE UNIVERSAL IDENTITY	9
APPLICATION WITH FOURTH ORDER ELASTIC CONSTANTS	10
The Velocities	10
The Equation of State	12
Numerical Results and Discussion	13
COMPARISON WITH AVAILABLE ELASTIC CONSTANTS	19
CONCLUSIONS	22
REFERENCES	24

## LIST OF TABLES

1. ELASTIC CONSTANTS ( $\times 10^{11}$ DYNES/CM <sup>2</sup> )	15
2. THIRD ORDER CONSTANTS FROM FOUR DIFFERENT EXPERIMENTS AS GIVEN BY BARSCH CONVERTED TO OUR FORMULATION	21

## LIST OF ILLUSTRATIONS

1. Schematic of the experimental acoustic measuring apparatus.	3
2. Configuration of system undergoing deformation.	3
3. $v_p, v_s$ as a function of pressure taking into consideration second order, third order, and fourth order elastic constants and comparing with experimental results.	16



	<u>Page</u>
4. P vs $\lambda_1$ is plotted and inclusion of the fourth order is found to be necessary to fit the equation of state.	17

## INTRODUCTION

Ultrasonic measurements at very high pressures give us the unique opportunity of obtaining the elastic properties of crystalline solids experiencing extremely large elastic deformations, which cannot be attained by other more conventional techniques. At best conventional stress-strain measurements in crystalline solids indicate elastic strains of the order of one percent; in sodium chloride the linear strain at a hydrostatic pressure of 270 kbar is nearly eleven percent.

Methods of continuum mechanics brought to bear on high pressure problems were inspired by Bridgeman's pioneering experimental work. Murnaghan<sup>2</sup> did the early theoretical work, inventing the finite strain method and using it to third order elastic constants for the equation of state. Birch<sup>3</sup> was another early worker using a somewhat different though valid formalism. A treatment giving the fourth order elastic constants for single crystals using lattice dynamics was given by Ghate<sup>4</sup>. The present treatment derives both the equation of state and the velocity in terms of the fourth order elastic constants by methods of continuum mechanics.

The methods used here are based on the tensor formalism developed in Green and Zerna.<sup>1</sup>

<sup>1</sup>Green, A. E., W. Zerna, Theoretical Elasticity, Oxford at the Clarendon Press, 1954.

<sup>2</sup>Murnaghan, F. D., Finite Deformations of an Elastic Solid, Dover Publications, Inc., New York.

<sup>3</sup>Birch, F., "The Effect of Pressure Upon the Elastic Parameters of Isotropic Solids, According to Murnaghan's Theory of Finite Strain," Jour. Appl. Phys. 9, 279, 1938.

<sup>4</sup>Ghate, P. B., "Fourth Order Elastic Coefficients," Jour. Appl. Phys. 35.2, 337, 1964.



The experimental techniques for obtaining high "quasi-hydrostatic" pressures<sup>5</sup> and for making high pressure ultrasonic interferometry measurements<sup>6</sup> were described in detail elsewhere and are shown schematically in Figure 1.

The direct experimental data obtained from ultrasonic interferometry consists of the frequency intervals  $\Delta f_p$  and  $\Delta f_s$  (for dilatational and shear modes respectively) for maximum destructive interference of the first echo of the ultrasonic echo train as observed on the cathode ray tube. Here the respective velocities are given by  $(\Delta f)/(2d)$  where  $d$  is the specimen thickness. The isotropic velocities were obtained through the use of the Decker equation and the frequency intervals in the universal identity relating the adiabatic density derivative of the pressure and the dilatational and shear isotropic velocities  $V_p$  and  $V_s$  respectively at all hydrostatic pressures:  $\partial p / \partial \rho_{ad} = V_p^2 - (4/3)V_s^2$ .

#### DERIVATION OF VELOCITIES FOR THE THEORY OF SMALL DEFORMATION SUPERPOSED ON FINITE UNIFORM DEFORMATION

Using the notation of Green and Zerna<sup>1</sup> and conforming to the experimental situation we apply a uniform hydrostatic deformation to the unstrained body  $B_0$  which becomes  $B$  in the new (pressurized) state. To the body  $B$  we apply an infinitesimal deformation (the ultrasonic wave) at time  $t$ , and represent this configuration as  $B'$  (Figure 2).

<sup>1</sup>Green, A. E., W. Zerna, Theoretical Elasticity, Oxford at the Clarendon Press, 1954.

<sup>5</sup>Kendall, D. P., P. V. Dembowski, T. E. Davidson, Rev. Sci. Inst. Vol. 46, No. 5 (1975).

<sup>6</sup>Frankel, J., J. F. Rich, C. G. Homan, J. Geoph. Res. Vol. 81, No. 35, 6357-6363, 1976.



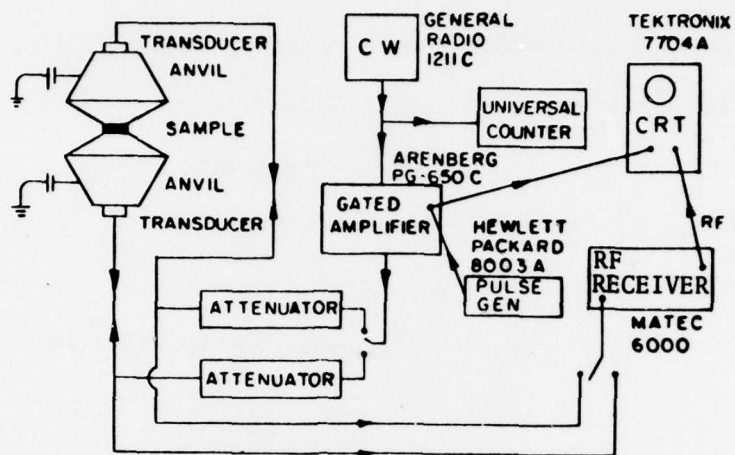


Figure 1. Schematic of the experimental acoustic measuring apparatus.

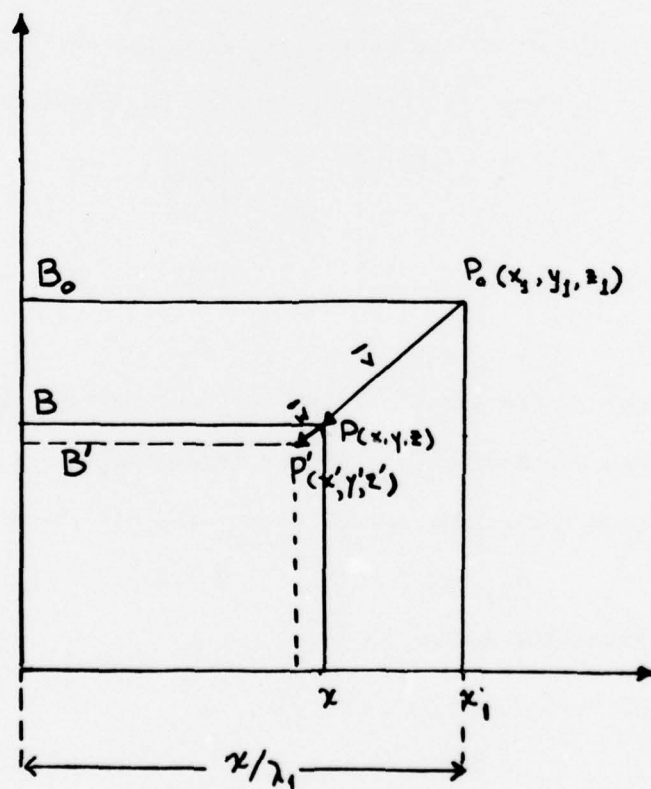


Figure 2. Configuration of system undergoing deformation.

Taking the moving coordinates to coincide with a fixed rectangular system  $(x, y, z)$  in the strained body B (Eulerian formulation) we have

$$\Theta_1 = x \quad \Theta_2 = y \quad \Theta_3 = z \quad (\text{strained body}) \quad (1)$$

The rectangular axes which define points in  $B_0$  are taken to coincide with the axes  $(x_1, x_2, x_3)$ .

The constant extension ratio  $\lambda_1$ , (the strain parameter) is given by

$$\lambda_1 = \frac{x}{x_1} = \frac{y}{y_1} = \frac{z}{z_1} \quad (2)$$

By taking the body (working) coordinates in the deformed body (i.e. the working coordinates move with the mass points of the body) and using the definition of the metric  $g_{ij}$  from the equation  $ds^2 = g_{ij} d\theta^i d\theta^j$  we have the metric tensor of the undeformed body

$$g_{ij} = \begin{bmatrix} 1/\lambda_1^2 & 0 & 0 \\ 0 & 1/\lambda_1^2 & 0 \\ 0 & 0 & 1/\lambda_1^2 \end{bmatrix}; \quad g^{ij} = \begin{bmatrix} \lambda_1^2 & 0 & 0 \\ 0 & \lambda_1^2 & 0 \\ 0 & 0 & \lambda_1^2 \end{bmatrix} \quad (3)$$

$$\text{and } g = 1/\lambda_1^6$$

Here the sub or superscripts refer to the covariant or contravariant tensor representations.  $g$  is the determinant of  $g_{ij}$ . In the same representation the metric tensor  $G_{ij}$  of the deformed body B is

$$G_{ij} = G^{ij} = \delta_{ij} \quad ; \quad G = 1 \quad (4)$$

where  $\delta$  is the Kronecker delta.

### The Deformed State (Under Finite Deformation)

The strain invariants are

$$I_1 = 3\lambda_1^2 \quad I_2 = 3\lambda_1^4 \quad I_3 = \lambda_1^6 \quad (5)$$

and the strain tensor elements become

$$\gamma_{ij} = \frac{1}{2} (G_{ij} - g_{ij}) = \frac{1}{2} \delta_{ij} (1 - \frac{1}{\lambda_1^2}) \quad (6)$$

The strain invariants (Eq. 5) are independent of the coordinates, they are constant for a given deformation. At this point we invoke Murnaghan's theorem (p. 61 of reference 2) for isotropic elastic media: "A deformable medium is isotropic if, and only if, the strain energy density is a function of the three strain invariants  $I_1, I_2, I_3$ ".

We can now express the strain energy density  $W$  in terms of  $I_1, I_2, I_3$  and after some manipulation (as given in reference 2) and using the principle of virtual work we can obtain the components of the stress tensor of the body under finite deformation.

$$\begin{aligned} \tau^{11} &= \phi \lambda_1^2 + 2\psi \lambda_1^4 + p \\ \tau^{22} &= \tau^{33} = \tau^{11} \\ \tau^{12} &= \tau^{13} = \tau^{23} = 0 \end{aligned} \quad (7)$$

$$\text{where } \phi = \frac{2}{\sqrt{I_3}} \frac{\partial W}{\partial I_1} \quad \psi = \frac{2}{\sqrt{I_3}} \frac{\partial W}{\partial I_2} \quad p = 2\sqrt{I_3} \frac{\partial W}{\partial I_3} \quad (8)$$

It should be noted here that since  $G_{ij} = \delta_{ij}$  the unit reference area is taken in the deformed coordinates. Equations (7) therefore represent the physical components of the stresses and in particular  $\tau^{11}$  is the hydrostatic pressure in our experiment ( $\tau^{11} = -p$ ).

<sup>2</sup>Murnaghan, F. D., Finite Deformations of an Elastic Solid, Dover Publications Inc., New York.

### The Perturbed State

We are now in a position to derive similar quantities for the perturbed case  $B'$ , the displacements due to the ultrasonic wave. In this new configuration the displacements can be written as the sum of the initial uniform displacement of the body  $\vec{v}$  and that of the perturbation  $\vec{w}$ ,  $\vec{v}(\theta_1, \theta_2, \theta_3, t) + \vec{w}(\theta_1, \theta_2, \theta_3, t)$  where  $\epsilon$  is a constant so small that the squares of  $\epsilon$  and its higher powers may be neglected compared to  $\epsilon$ .

The covariant base vectors of the coordinate system  $\theta_i$  at points  $P'$  of  $B'$  are denoted by  $\vec{G}_i + \epsilon \vec{G}_i' = \vec{r}_{,i} + \vec{v}_{,i} + \epsilon \vec{w}_{,i}$  where  $\vec{r}$  is the location of point  $P_0$ , and the commas indicate differentiation with respect to the  $i^{\text{th}}$  coordinate.

To the first order in  $\epsilon$  the perturbed metric tensor  $G'_{ij}$  (the perturbed state is usually denoted by primed quantities) of the deformed body referred to base vectors of body  $B$  is:

$$G'_{ij} = \begin{bmatrix} 2 \frac{\partial u}{\partial x} & \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} & \frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} \\ (\text{symm}) & 2 \frac{\partial v}{\partial y} & \frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} \\ & & 2 \frac{\partial w}{\partial z} \end{bmatrix} = - G^{ij'} \quad (9)$$

$$w = \omega_m \vec{G}_m = \omega^m \vec{G}_m$$

$$\text{and } \omega_1 = u \quad \omega_2 = v \quad \omega_3 = w$$

The perturbation in the strain invariants is

$$\begin{aligned} I'_1 &= 2\lambda_1^2 \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \\ I'_2 &= 4\lambda_1^4 \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \\ I'_3 &= 2\lambda_1^6 \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \end{aligned} \quad (10)$$

The perturbation components of the stress tensor are given by (see ref. [1] p. 119).

$$\begin{aligned} \tau'^{11} &= C_{11} \frac{\partial u}{\partial x} + C_{12} \left( \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \\ \tau'^{22} &= C_{11} \frac{\partial v}{\partial y} + C_{12} \left( \frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} \right) \\ \tau'^{33} &= C_{11} \frac{\partial w}{\partial z} + C_{12} \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \end{aligned} \quad (11)$$

$$\begin{aligned} \tau'^{12} &= C_{44} \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \\ \tau'^{23} &= C_{44} \left( \frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} \right) \\ \tau'^{31} &= C_{44} \left( \frac{\partial w}{\partial x} + \frac{\partial u}{\partial z} \right) \end{aligned} \quad (12)$$

Where

$$\begin{aligned} C_{11} &= -\tau'^{11} + 2A\lambda_1^4 + 8B\lambda_1^8 + 2C\lambda_1^{12} + 8D\lambda_1^{10} + 4E\lambda_1^8 + 8F\lambda_1^6 \\ C_{12} &= -\lambda_1^2 \phi + p + 2A\lambda_1^4 + 8B\lambda_1^8 + 2C\lambda_1^{12} + 8D\lambda_1^{10} + 4E\lambda_1^8 + 8F\lambda_1^6 \\ C_{44} &= \frac{1}{2}(C_{11} - C_{12}) \end{aligned} \quad (13)$$

And

$$\begin{aligned} A &= \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_1^2}, \quad B = \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_2^2}, \quad C = \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_3^2} \\ D &= \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_2 \partial I_3}, \quad E = \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_3 \partial I_1}, \quad F = \frac{2}{\sqrt{I_3}} \frac{\partial^2 W}{\partial I_1 \partial I_2} \end{aligned} \quad (14)$$

<sup>1</sup>Green, A. E., W. Zerna, Theoretical Elasticity, Oxford at the Clarendon Press, 1954.



The physical components of perturbation in stresses,  $t'^{rs}$ , can be obtained by the contravariant transformation rule, and to the first order in  $\epsilon$  and referring to the  $B'$  configuration we have

$$t'^{rs} = \tau'^{rs} + \tau^{ms} \frac{\partial \omega_r}{\partial \theta^m} + \tau^{rm} \frac{\partial \omega_s}{\partial \theta^m} \quad (15)$$

Using (12) we have

$$\begin{aligned} t'^{11} &= (C_{11} + 2\tau^{11}) \frac{\partial u}{\partial x} + C_{12} \left( \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \\ t'^{22} &= (C_{11} + 2\tau^{11}) \frac{\partial v}{\partial y} + C_{12} \left( \frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} \right) \\ t'^{33} &= (C_{11} + 2\tau^{11}) \frac{\partial w}{\partial z} + C_{12} \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \\ t'^{12} &= (C_{44} + \tau^{11}) \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \\ t'^{23} &= (C_{44} + \tau^{11}) \left( \frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} \right) \\ t'^{31} &= (C_{44} + \tau^{11}) \left( \frac{\partial w}{\partial x} + \frac{\partial u}{\partial z} \right) \end{aligned} \quad (16)$$

The equation of motion, with  $f'^r$  as acceleration components, become

$$\frac{\partial t'^{rs}}{\partial \theta^s} = \rho f'^r \quad (17)$$

From (17) and (16) we have (after some manipulation)

$$\begin{aligned} \frac{1}{2} (C_{11} - C_{12} + 2\tau^{11}) \nabla^2 u + \frac{1}{2} (C_{11} + C_{12} + 2\tau^{11}) \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 v}{\partial x \partial y} + \frac{\partial^2 w}{\partial x \partial z} \right) &= \rho \frac{\partial^2 u}{\partial t^2} \\ \frac{1}{2} (C_{11} - C_{12} + 2\tau^{11}) \nabla^2 v + \frac{1}{2} (C_{11} + C_{12} + 2\tau^{11}) \left( \frac{\partial^2 v}{\partial y^2} + \frac{\partial^2 w}{\partial y \partial z} + \frac{\partial^2 u}{\partial x \partial y} \right) &= \rho \frac{\partial^2 v}{\partial t^2} \\ \frac{1}{2} (C_{11} - C_{12} + 2\tau^{11}) \nabla^2 w + \frac{1}{2} (C_{11} + C_{12} + 2\tau^{11}) \left( \frac{\partial^2 w}{\partial z^2} + \frac{\partial^2 u}{\partial x \partial z} + \frac{\partial^2 v}{\partial y \partial z} \right) &= \rho \frac{\partial^2 w}{\partial t^2} \end{aligned} \quad (18)$$

Vectorially adding above with  $\vec{w} = \vec{i}u + \vec{j}v + \vec{k}w$  we have

$$\frac{1}{2} (C_{11} - C_{12} + 2\tau^{11}) \nabla^2 \bar{w} + \frac{1}{2} (C_{11} + C_{12} + 2\tau^{11}) \nabla(\nabla \cdot \bar{w}) = \rho \ddot{\bar{w}} \quad (19)$$

This is the equation of motion of body B' in terms of perturbation ( $\bar{w}$ ) of displacement of body B.

Taking the divergence and curl of (19) we have

$$\begin{aligned} \nabla^2 (\nabla \cdot \bar{w}) &= \left( \frac{\rho}{C_{11} + 2\tau^{11}} \right) \frac{\partial^2}{\partial t^2} (\nabla \cdot \bar{w}) \\ \nabla^2 (\nabla \times \bar{w}) &= \left( \frac{2\rho}{C_{11} - C_{12} + 2\tau^{11}} \right) \frac{\partial^2}{\partial t^2} (\nabla \times \bar{w}) \end{aligned} \quad (20)$$

#### THE UNIVERSAL IDENTITY

From (16) we have

$$t^{,11} + t^{,22} + t^{,33} = (C_{11} + 2C_{12} + 2\tau^{11}) \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} \right) \quad (21)$$

$$\text{let } t^{,11} = t^{,22} = t^{,33} = -\Delta \text{ (Pressure)} = -\Delta P \quad (22)$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = \frac{-\Delta \text{ (Volume)}}{\text{Unperturbed Volume}} = -\Delta \xi$$

$$\text{hence } \frac{\Delta \rho}{\rho} = -\Delta \xi \quad (23)$$

(23) and (22) with (21) give

$$\rho \frac{dP}{d\rho} = \text{Limit}_{\Delta \xi \rightarrow 0} \quad \rho \frac{\Delta P}{\Delta \rho} = \frac{1}{3} (C_{11} + 2C_{12} + 2\tau^{11}) \quad (24)$$

Hence from (24) and (20) eliminating  $C_{11}$ ,  $C_{12}$ ,  $\tau^{11}$  we have

$$V_p^2 = \frac{4}{3} V_s^2 + \frac{dP}{d\rho} \quad (25)$$

(25) is the universal identity<sup>7</sup>.

<sup>7</sup>Truesdell, C., Handbuch der Physik, Ed.S. Flugge, Vol III/3, Springer Verlag, Heidelberg, New York.

From (20) we have the velocities

$$\begin{aligned} v_p &= \left( \frac{C_{11} + 2\tau^{11}}{\rho} \right)^{1/2} && \text{Dilatational Velocity} \\ v_s &= \left( \frac{C_{11} - C_{12} + 2\tau^{11}}{2\rho} \right)^{1/2} && \text{Shear Velocity} \end{aligned} \quad (26)$$

Where  $C_{11}$ ,  $\tau^{11}$  etc. can be computed in terms of derivatives of  $W$  with respect to strain invariants from (13), (14) and (8).

#### APPLICATION WITH FOURTH ORDER ELASTIC CONSTANTS

##### The Velocities

In this section we assume that the elastic strain energy for the material under consideration can be expanded up to fourth order terms in strain invariants. The strain invariants defined by Murnaghan<sup>2</sup> are slightly different than those of Green and Zerna<sup>1</sup>. For the sake of easy comparison we give the transformation, denoting those of Murnaghan by a superscript  $c$ ,

$$\begin{aligned} I_1^c &= \frac{1}{2} (I_1 - 3) \\ I_2^c &= \frac{1}{4} [(I_2 - 3) - 2(I_1 - 3)] \\ I_3^c &= \frac{1}{8} [(I_3 - 1) + (I_1 - 3) - (I_2 - 3)] \end{aligned} \quad (27)$$

$$\begin{aligned} W &= \frac{1}{2}(\lambda + 2\mu) I_1^{c^2} - 2\mu I_2^c + \frac{(\lambda + 2\mu)}{3} I_1^{c^3} - 2m I_1^c I_2^c + n I_3^c \\ &+ 16q(I_1^c)^4 + 16r(I_1^c)^2(I_2^c) + 16s(I_1^c I_3^c) + 16t(I_2^c)^2 \end{aligned} \quad (28)$$

<sup>1</sup>Green, A. E., W. Zerna, Theoretical Elasticity, Oxford at the Clarendon Press, 1954.

<sup>2</sup>Murnaghan, F. D., Finite Deformations of an Elastic Solid, Dover Publications Inc., New York.

Where  $\lambda, \mu$       Lamé's second order elastic constants  
 $\ell, m, n$       (Murnaghan's) third order elastic constants  
 $q, r, s, t$       Fourth order elastic constants  
(Factor 16 is for convenience only)

Substituting equation (27) into (28) we have

$$\begin{aligned}
W = & \lambda \frac{1}{8} (I_1 - 3)^2 \\
& + \mu \frac{1}{4} [(I_1 - 3)^2 - 2(I_2 - 3) + 4(I_1 - 3)] \\
& + \ell \frac{1}{24} (I_1 - 3)^3 + m \frac{1}{12} [(I_1 - 3)^3 - 3(I_1 - 3)(I_2 - 3) + 6(I_1 - 3)^2] \\
& + n \frac{1}{8} [(I_3 - 1) + (I_1 - 3) - (I_2 - 3)] \\
& + q(I_1 - 3)^4 + r[(I_1 - 3)^2(I_2 - 3) - 2(I_1 - 3)^3] \\
& + s[(I_1 - 3)(I_3 - 1) + (I_1 - 3)^2 - (I_1 - 3)(I_2 - 3)] \\
& + t[(I_2 - 3)^2 - 4(I_2 - 3)(I_1 - 3) + 4(I_1 - 3)^2]
\end{aligned} \tag{29}$$

Using (26), (13) and (5) and some manipulation we have

$$\begin{aligned}
\rho V_p^2 = & \lambda \left( \frac{5\lambda_1}{2} - \frac{3}{2\lambda_1} \right) + \mu \left( 3\lambda_1 - \frac{1}{\lambda_1} \right) + 3\ell \left( \frac{7\lambda_1^3}{4} - \frac{5\lambda_1}{2} + \frac{3}{4\lambda_1} \right) \\
& + 2m \lambda_1 (\lambda_1^2 - 1) + \frac{n}{4\lambda_1} (\lambda_1^2 - 1)^2 + 216q (\lambda_1^2 - 1)^2 \left( 3\lambda_1 - \frac{1}{\lambda_1} \right)
\end{aligned} \tag{30}$$

$$+ 24r (\lambda_1^2 - 1)^2 \left( 8\lambda_1 - \frac{3}{\lambda_1} \right) + 8s (\lambda_1^2 - 1)^2 \left( 2\lambda_1 - \frac{1}{\lambda_1} \right) + 8t (\lambda_1^2 - 1)^2 \left( 7\lambda_1 - \frac{3}{\lambda_1} \right)$$

and

$$\begin{aligned}
\rho V_s^2 = & \frac{3\lambda}{2} \left( \lambda_1 - \frac{1}{\lambda_1} \right) + \mu \left( 2\lambda_1 - \frac{1}{\lambda_1} \right) + \frac{9\ell}{4\lambda_1} (\lambda_1^2 - 1)^2 + \frac{3m\lambda_1}{2} (\lambda_1^2 - 1) \\
& + \frac{n}{4\lambda_1} (\lambda_1^2 - 1) + 216q \frac{1}{\lambda_1} (\lambda_1^2 - 1)^3 + 18r (\lambda_1^2 - 1)^2 \left( 3\lambda_1 - \frac{4}{\lambda_1} \right) \\
& + 2s (\lambda_1^2 - 1)^2 \left( \lambda_1 - \frac{4}{\lambda_1} \right) + 12t (\lambda_1^2 - 1) \left( \lambda_1 - \frac{2}{\lambda_1} \right)
\end{aligned} \tag{31}$$

In the above expression there are two second order, three third order and four fourth order elastic constants. However, only two unique combinations of each third and fourth order elastic constants appear in (30) and (31); these combinations can be easily found by collecting equal powers of the strain parameter  $\lambda_1$ . These are

$$\begin{aligned}\alpha &= (9\ell+n) \\ \beta &= (3\ell+2m)\end{aligned}\quad \begin{array}{l} \text{Third Order} \\ \text{Fourth Order} \end{array}\quad (32)$$

$$\begin{aligned}\gamma &= (27q + 9r + s + 3t) \\ \delta &= (81q + 24r + 2s + 7t)\end{aligned}$$

Using (32) the expressions in (30) and (31) become

$$\begin{aligned}V_p^2 \rho &= \lambda_1^5 (8\delta) + \lambda_1^3 \left( \frac{1}{4} \alpha + \beta - 8\gamma - 16\delta \right) + \lambda_1 \left( \frac{5\lambda}{2} + 3\mu - \frac{\alpha}{2} - \beta + 16\gamma + 8\delta \right) \\ &\quad + \frac{1}{\lambda_1} \left( -\frac{3\lambda}{2} - \mu + \frac{1}{4} \alpha - 8\gamma \right)\end{aligned}\quad (33)$$

$$\begin{aligned}V_s^2 \rho &= \lambda_1^5 (6\delta - 10\gamma) + \lambda_1^3 \left( \frac{3}{4} \beta - 12\delta + 12\gamma \right) + \lambda_1 \left( \frac{3\lambda}{2} + 2\mu - \frac{\alpha}{4} - \frac{3}{4} \beta \right) \\ &\quad + 6\gamma + 6\delta + \frac{1}{\lambda_1} \left( -\frac{3\lambda}{2} - \mu + \frac{1}{4} \alpha - 8\gamma \right)\end{aligned}\quad (34)$$

#### The Equation of State

Using (7) and noting that  $\tau^{11} = -\text{Pressure}$  we have the equation

$$-P = \frac{1}{2} (3\lambda + 2\mu) \frac{1}{\lambda_1} (\lambda_1^2 - 1) + \alpha \frac{1}{4\lambda_1} (\lambda_1^2 - 1)^2 + \frac{8\gamma}{\lambda_1} (\lambda_1^2 - 1)^3 \quad (35)$$

The first two terms on right side of (35) represent Murnaghan's equation, the additional term is due to fourth order elastic constants, and plays an important role at high strains.



It should be noted that in Equation (35) only one combination of each order of elastic constants appears. This is the consequence of the universal identity which has to be satisfied, independently of the assumed elastic potential. This phenomenon has far reaching consequences, as will be explained in the sequel, in obtaining the equation of state, purely from the velocity ratio.

#### Numerical Results and Discussion

As explained in reference 6,  $\Delta f_p$  and  $\Delta f_s$  are obtained experimentally and are related to  $V_p$  and  $V_s$  by  $\Delta f_p = V_p/2d$   $\Delta f_s = V_s/2d$  where  $d$  is the specimen thickness. A spline function fit was made to these  $\Delta f_p$  and  $\Delta f_s$  and using the universal identity (25) (which is independent of the elastic constants used) and Decker's equation of state<sup>8</sup>,  $d$  was determined from

$$d = \left[ -\frac{\lambda_1^4}{12\rho_0} \frac{dp/d\lambda_1(1+\Delta)}{(\Delta f_p^2 - 4/3\Delta f_s^2)} \right]^{1/2}, \quad (1+\Delta = 1.054) \quad (36)$$

and  $V_p, V_s$  computed for all pressures.

The Frankel, Rich, and Homan<sup>6</sup> (FRH) and Voronov and Grigorev<sup>9</sup> (VG) velocity data were both analyzed. Since the FRH data starts at 25 kbar (the 25 kbar point having been determined by matching velocity ratios of FRH to VG at 25 kbar) only the VG velocity and  $\partial v/\partial p$  were used near zero pressure to give the second order elastic constants  $\lambda$  and  $\mu$ , and the third order elastic constants  $\alpha$  and  $\beta$ . Having obtained the second

<sup>6</sup>Frankel, J., J. F. Rich, C. G. Homan, J. Geoph. Res. Vol. 81, No. 35, 6357-6363, 1976.

<sup>8</sup>Decker, D. L., W. A. Barrett, L. Merrill, H. T. Hall, and J. D. Barnett, "High Pressure Calibration: A Critical Review," J. Phys. Chem., Ref. Date 1, 773-835, 1972.

<sup>9</sup>Voronov, F. F., and Grigorev, S. B., "Influence of Pressures up to 100 Kbar on Elastic Properties of Silver Sodium and Cesium Chloride," Sov. Phys. Solid State, Vol. 18, No. 2, 325-328, 1976.

and third order elastic constants from VG alone the fourth order elastic constants were obtained in two ways, by analyzing the FRH data and the VG velocity data. In addition to that, one of the fourth order elastic constants  $\gamma$ , was obtained by fitting the results of the Decker equation to our formulation of the equation of state (Eq. 35). For the ultrasonic data the fourth order elastic constants  $\gamma$  and  $\delta$  were computed by minimizing the integral of the sum of the squares of the differences between the experimental values of  $v_p$  and  $v_s$  and the theoretical expressions over the parameters  $\gamma$  and  $\delta$ . The results are given in Table 1.

In Figure 3 we plot  $v_p$  and  $v_s$  as a function of pressure for inclusion of second, third and fourth order elastic constants. The experimental results are also plotted.

An interesting observation from Figure 3 is the peak in the shear velocity as predicted by VG data using fourth order elastic constants. These elastic constants predict a shear velocity which peaks at about 80 kbar. A peak would be interesting from lattice dynamics considerations using the Born criterion of phase changes.<sup>6</sup> The FRH experimental data does not show such a peak.

In terms of our formulation the results from the VG data agree very well with the Decker equation (see  $\gamma$  in Table 1 and Figure 4). The fourth order elastic constants from VG evaluated up to 80 kbar

<sup>6</sup>Frankel, J., J. F. Rich, C. G. Homan, J. Geoph. Res. Vol. 81, No. 35, 6357-6363, 1976.

TABLE 1. Elastic constants obtained in our formulation. The second and third order constants were obtained from low pressure velocity data of Voronov and Grigorev<sup>9</sup>. The fourth order elastic constants are obtained from present analysis based on velocities reported in ref. 6. The fourth order constant under source D was obtained from the Decker equation using VG values for second and third order elastic constants.

ELASTIC CONSTANTS ( $\times 10^{11}$ DYNES/CM <sup>2</sup> )						
Source	Second Order		Third Order		Fourth Order	
	$\lambda$	$\mu$	$\alpha$	$\beta$	$\gamma$	$\delta$
VG	1.5430	1.4699	-54.8724	-30.845	4.4701	18.3613
FRH	"	"	"	"	6.5206	28.1030
D	"	"	"	-	4.8716	-

<sup>6</sup>Frankel, J., J. F. Rich, C. G. Homan, J. Geoph. Res. Vol. 81, No. 35, 6357-6363, 1976.

<sup>9</sup>Voronov, F. F., and Grigorev, S. B., "Influence of Pressures up to 100 Kbar on Elastic Properties of Silver Sodium and Cesium Chloride," Sov. Phys. Solid State, Vol. 18, No. 2, 325-328, 1976.

## Velocity Comparison

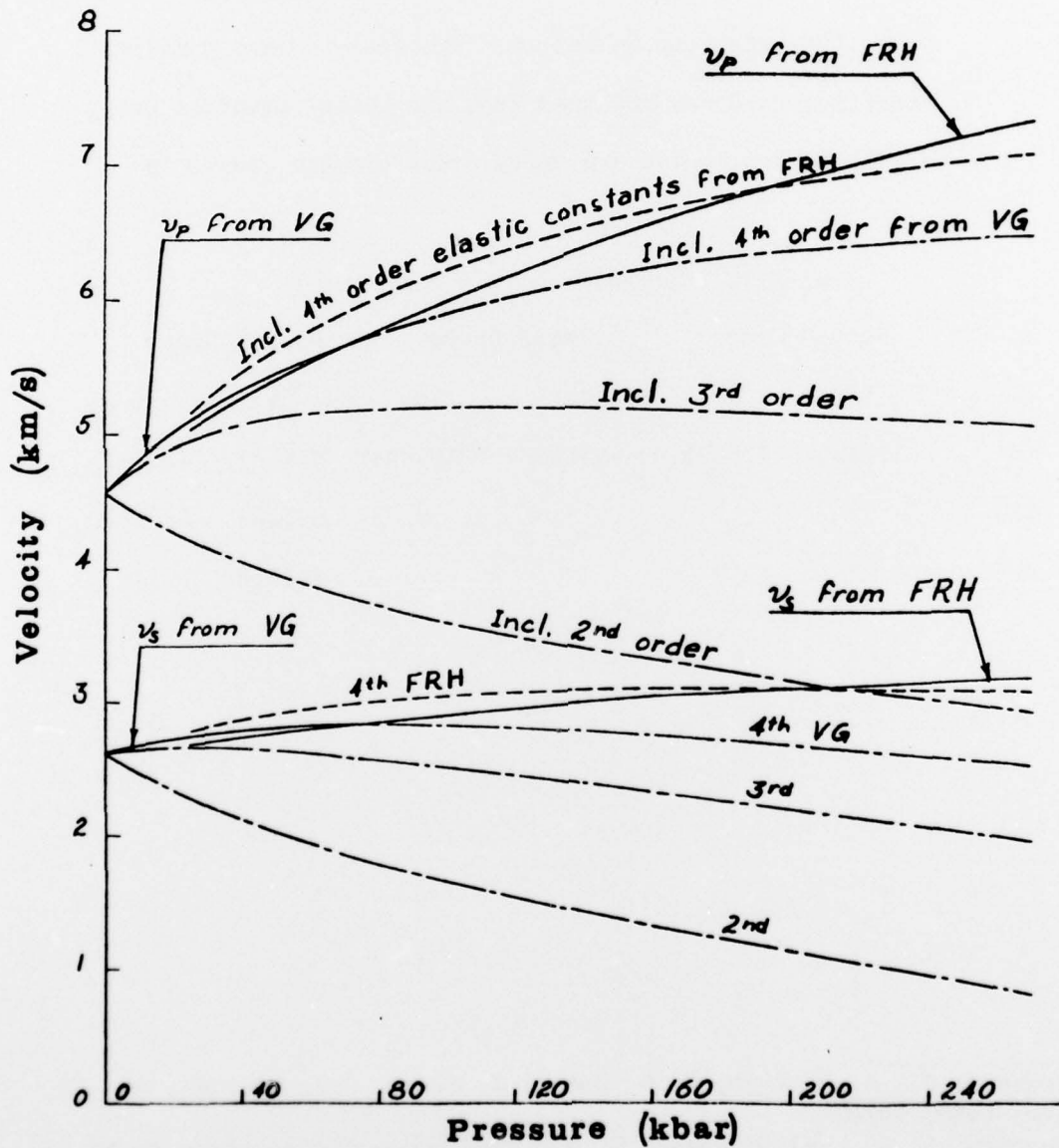


Figure 3.  $v_p, v_s$  as a function of pressure taking into consideration second order, third order, and fourth order elastic constants and comparing with experimental results.

# Equation of State Comparison

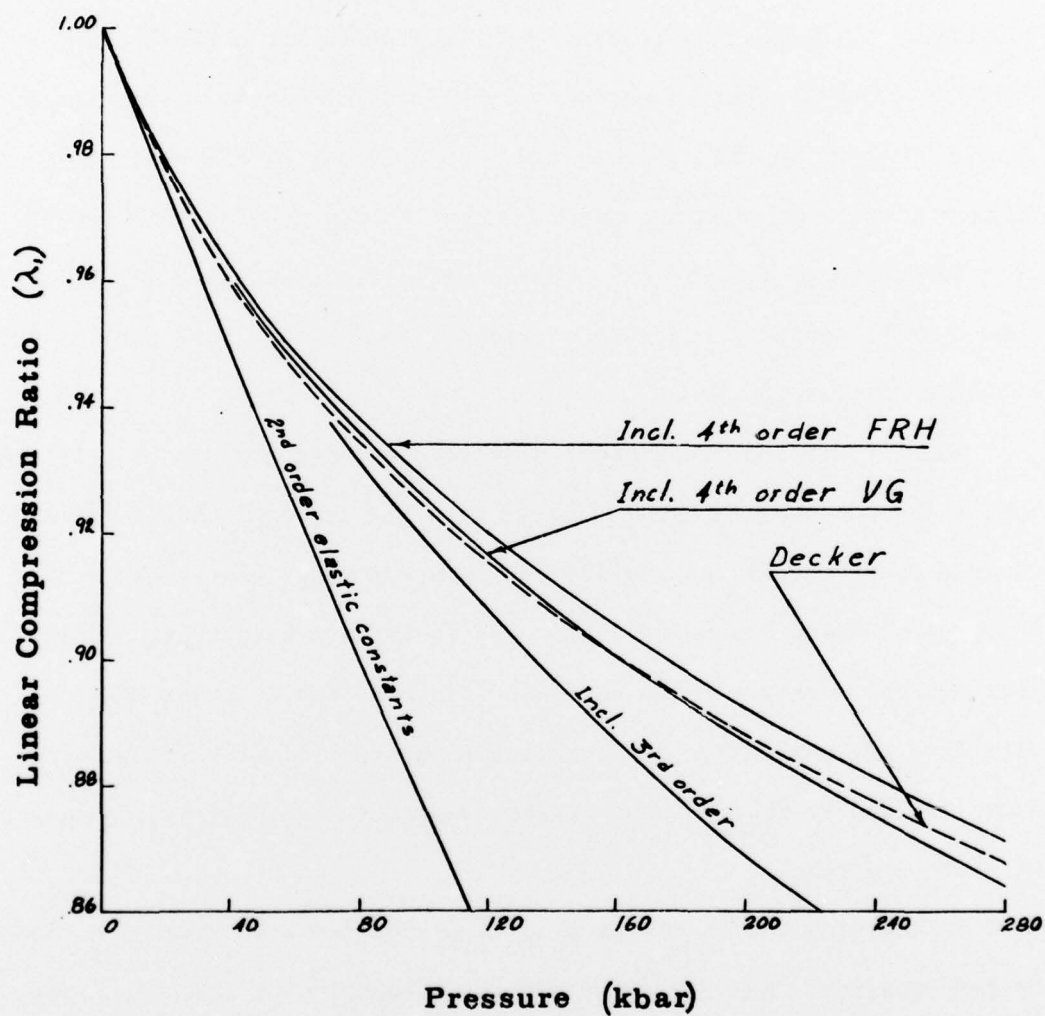


Figure 4.  $P$  vs  $\lambda_1$  is plotted and inclusion of the fourth order term is found to be necessary to fit the equation of state.



predict the equation of state to 270 kbar. The equation of state obtained from the FRH velocities is also included in Figure 4 as an internal check since the Decker equation was used to obtain the velocities. The results from the FRH data differ from the Decker equation curve by about 5 percent of the change in the linear dimension to 270 kbar, or on the pressure scale by  $\approx 10$  out of 270 kbar (4%). Figures 3 and 4 point up an anomaly: even though the fourth order elastic constants fit the FRH deduced velocities better to 270 kbar, the fourth order elastic constants obtained from VG to 80 kbar predict the equation of state better.

Further work on fifth order elastic constants and use of velocity ratios is indicated. Figures 3 and 4 strongly indicate that third order elastic constants are not sufficient in predicting velocities or the equation of state beyond 80 kbar. The fact that the present development not only derives the equation of state to fourth order elastic constants but enables us to determine them from velocity information will find application in very high pressure or high compression equation of state studies.

In deriving the FRH results of Figures 3 and 4 we have used the Decker equation. However, an independent equation of state can also be obtained if we have good velocity data in the neighborhood of zero pressure.

The computation is done as follows: a spline function fit is obtained to the available data near zero pressure (using interpolating splines) and the derivatives obtained at  $P = 0$  (i.e.  $\lambda_1 = 1$ ). From (33) and (34) we then obtain  $\lambda, \mu, \alpha, \beta$ , the second and third order elastic constants. With these we minimize the integral of the difference of the squares of the experimentally obtained  $\Delta f_p / \Delta f_s$ , with respect to  $\gamma$  and  $\delta$  (fourth order elastic constants), and the ratio of equations (33) and (34) over the required range of pressure (least square fit). Once  $\gamma$  and  $\delta$  are thus determined the equation of state is then easily obtained from equation (35). This method has an additional advantage, that it is not necessary to measure the deformed dimensions of the specimen.

#### COMPARISON WITH AVAILABLE ELASTIC CONSTANTS

The fourth order elastic constants are not yet available in the open literature. However, Barsch<sup>10</sup> has done an extensive review and analysis for third order isotropic elastic constants. Comparing our equation (29) to Barsch's equation 5 we get in his notation

$$\begin{aligned} n &= 7/2 C_{456} \\ m &= C_{144} - 7/4 C_{456} \\ l &= 1/4 (C_{123} + 2m - n) \end{aligned} \tag{37}$$

<sup>10</sup>Barsch, G. R. "Relations Between Third Order Elastic Constants of Single Crystals and Polycrystals," Jour. Appl. Phys., Vol. 39, 8, 3780, 1968.

We evaluated data given by Barsch<sup>10</sup> from four different sodium chloride experiments by substituting (37) for the third order elastic constants in (32). The results are given in Table 2. Since the NaCl 2 data falls strongly out of the pattern of the other three, we compared the  $\alpha$  and  $\beta$  as given in Table 1 with the average of NaCl 1, 3 and 4. The results are in excellent agreement.

Table 1 also shows the satisfactory agreement between the values of  $\gamma$  and  $\delta$  obtained from different experiments, and the Decker Equation, as discussed above.

No mention has been made so far as to the nature of the elastic constants obtained. We note two factors (a) the specimen in our experiment was compressed isothermally; (b) velocity measurements at the frequencies of our experiment ( $\sim 10\text{MHz}$ ) are adiabatic.

Hence in high pressure experiments we make adiabatic measurements on a state arrived at by isothermal deformation. (The hydrostatic pressurization is assumed thermodynamically reversible, which is not totally true for quasi-hydrostatic systems.) The elastic constants thus arrived at are known as mixed elastic constants in the literature. No corrections have been made from mixed to isothermal elastic constants because the difference for NaCl can be considered small enough to fall within the uncertainty of the determination.<sup>11</sup>

<sup>10</sup>Barsch, G. R. "Relations Between Third Order Elastic Constants of Single Crystals and Polycrystals," Jour. Appl. Phys., Vol. 39, 8, 3780, 1968.

<sup>11</sup>Barsch, G. R. and Z. P. Chang, "Adiabatic Isothermal and Intermediate Pressure Derivatives of the Elastic Constants from Cubic Symmetry," Phys. Stat. Sol. 19, 139 (1967), and private communication (Chang).

TABLE 2. THIRD ORDER CONSTANTS FROM FOUR DIFFERENT EXPERIMENTS  
AS GIVEN BY BARSCH<sup>10</sup> CONVERTED TO OUR FORMULATION

Comparison of Third Order Elastic Constants of NaCl ( $\times 10^{11}$  dynes/cm<sup>2</sup>)

					Average of 1,3,4	%Difference wrt Table 1
$\alpha$	-56.86	-33.53	-55.76	-51.18	-54.27	1.1
$\beta$	-32.33	-24.16	-32.22	-29.96	-30.76	0.27

<sup>10</sup>Barsch, G. R., "Relations Between Third Order Elastic Constants of Single Crystals and Polycrystals," Jour. Appl. Phys., Vol. 39, 8, 3780, 1968.



## CONCLUSIONS

1. Using fundamental principles we have derived the equation of state and velocities in terms of second, third, and fourth order elastic constants, for an isotropic solid.

2. The results imply that a) with sufficiently accurate velocity data near zero pressure and the velocity ratio measured to high pressures an equation of state can be determined outside of regions where phase changes take place, b) with sufficiently good velocity data near zero pressure or at low pressures a pressure scale can be predicted from experimentally obtained velocity ratio information.

3. The mechanism of computation can easily be extended to fifth order elastic constants.

4. In general wave methods are much more sensitive for the measurement of elastic properties of matter than static measurements because of relative sensitivity of velocity measurements to volume measurements at these pressures.

5. As a last point it is useful to stress the complete generality and the high pressure predictive powers of the method derived here together with the velocity ratio concept.

The equations are derived from first principles and apply to any isotropic solid in a pressure-volume range where no phase changes take place. These can be carried to fourth and fifth order elastic constants and the constants can be obtained to any pressure to which the velocity ratio can be measured. The equation of state derived from these principles with the fourth and fifth order elastic constants is



expected to have predictive powers to higher than the measured pressures better than any method heretofore developed.

This will be useful in obtaining the very high pressure molecular equation of state for materials such as rare gas solids. These molecular results will then be compared with available equations for the metallic state and the pressure of the molecular-metallic transition will then be predicted, together with its estimated uncertainty.

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